Attorney Docket: 2000DE402D
Serial No.: 10/668,005
Art Unit No.: 1714
Declaration under 37 C.F.R. 1.132

PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: Matthias KRULL et al. Docket: 2000DE402D
Serial No.: 10/668,005 Group Art Unit: 1714
Filed: September 22, 2003 Examiner: Toomer, C.
For: **MULTIFUNCTIONAL ADDITIVE FOR FUEL OILS**

DECLARATION UNDER 37 CFR 1.132

Mail Stop
Amendment
Commissioner for Patents
PO Box 1450
Alexandria, VA 22313-1450

Dear Sir:

I, Matthias Krull, state that I am a resident of Am Rheinhessenblick 27, D-55296 Harxheim, Federal Republic of Germany; that I am a citizen of the Federal Republic of Germany; that I am a chemist having earned the degree of Dr. rer. nat. (corresponds to Ph. D.) from the Free University Berlin, Federal Republic of Germany, in 1989.

I am acquainted with the subject matter of the above subject Application and I am one of the named inventors of Application No. 10/668,005, filing date of September 22, 2003 in the name of Matthias Krull et al. for "MULTIFUNCTIONAL ADDITIVE FOR FUEL OILS."

I have been employed for 16 years in the Research and Development department of Hoechst AG, Frankfurt, Germany, which was succeeded by Clariant GmbH, Frankfurt, Germany, where my work has focused on oilfield chemicals and especially on cold flow additives for mineral oils.

CERTIFICATE OF MAILING/TRANSMISSION (37 CFR 1.8a) and 1.10

I hereby certify that this correspondence is, on the date shown below, being transmitted by facsimile to the U.S. Patent and Trademark Office. (Fax No. (571) 273-8300 (8) pages.

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Additive combinations of amphiphiles and terpolymers were tested for their performance on CFPP and lubricity improvement in different ratios and in comparison to conventional EVA copolymers. Additives used were

Amphiphile 1 (glycerol monooleate) as described in the original application.
Amphiphile 6 (tall oil fatty acid) as described in the original application.
Polymer A (ethylene-vinyl acetate copolymer, Comparison), as described in the original application.
Polymer F (terpolymer containing 85.2 mol-% ethylene, 10.4 mol-% vinyl acetate and 4.4 mol.-% vinyl neodecanoate; melt viscosity V_{140} of 165 mPas)
Polymer G (terpolymer containing 87.0 mol-% ethylene, 9.4 mol-% vinyl acetate and 3.6 mol.-% vinyl neoundecanoate; melt viscosity V_{140} of 151 mPas)
Polymer H (terpolymer containing 83.3 mol-% ethylene, 11,4 mol-% vinyl acetate and 6.3 mol.-% vinyl neononanoate; melt viscosity V_{140} of 187 mPas)

Solubility behavior according to British Rail test, lubricity activity according to the HFRR test and CFPP according to EN 116 were determined in the same manner as described in the subject application. The solubility behavior of the additives was determined according to the British Rail test, as follows: 400 ppm of a dispersion of the additive combination, heated to 22°C, are metered into 200 ml of the test oil heated to 22°C (cf. Table 3) and shaken vigorously for 30 seconds. After storage for 24 hours at +3°C, shaking was carried out again for 15 seconds and filtration was then carried out at +3°C in three portions of 50 ml each over a 1.6 µm glass fiber microfilter (Ø 25 mm; Whatman GFA, Order No. 1820025). The ADT value is calculated from the three filtration times T_1, T_2, and T_3, as follows:

$$ADT = \frac{(T_3 - T_1)}{T_2} \cdot 50$$

An ADT value of < 15 is regarded as an indication that the gas oil can be satisfactorily used in normally cold weather. Products having ADT values of > 25 are considered not to be filterable.

The lubricating activity of the additives was determined by means of an HFRR apparatus from PCS Instruments. The additives heated to 22°C are metered into the oil heated to 22°C and are shaken vigorously for 30 seconds. After storage for 25 hours at +3°C, the oil is filtered according to the conditions of the British Rail test and the lubricating activity is determined for the filtrate in the HFRR test. The high frequency reciprocating rig test (HFRR) is described in D. Wei, H. Spikes, Wear, Vol. 111, No. 2, p. 217, 1986 and is carried out at 60°C. The results are stated as a coefficient of friction and a wear scar (WSD). A low coefficient of friction and a low wear scar indicate good lubricating activity.

The cold flow improver polymers A and F to H were applied as 50 % active dispersions in organic solvent. They were mixed with the amphiphiles prior to testing in the fuels.

Tests were performed in two fuels (test oils 6 and 7) with the following characteristics:

		Test oil 6	Test oil 7
Cloud Point (CP)	[°C]	-7,5	-9,9
Cold Filter Plugging Point (CFPP)	[°C]	-9	-10
n-Paraffin content	[wt.-%]	19	21
Initial boiling point	[°C]	185	176
Boiling range 90-20%	[K]	96	93
FBP-90%	[K]	30	20
Final boiling point (FBP)	[K]	361	348
Density	[g/cm³]	0,836	0,835
S content	[ppm]	15	6
HFRR-WSD	[µm]	674	724
Average differential time (ADT)		5,4	4,8

Table A, which shows the test results obtained using test oil 6, provides experimental data obtained with additive combinations containing a <u>major amount of amphiphile</u>

and a minor amount of cold flow improver (CFI). The results for both Polymer F and for Polymer G show that an additive combination containing 35 ppm of CFI (23 % of total amount of CFI and amphiphile) and 120 ppm of amphiphile 6 (77 % of total amount of CFI and amphiphile) gives rise to a CFPP improvement in comparison to the treatment with CFI alone. This effect as well as an unchanged filterability (as measured by ADT) was observed only when terpolymers: Polymer F and Polymer G made of ethylene, vinyl acetate and the vinyl ester of a neocarboxylic aicd were used, but not with the comparative EVA copolymer A.

Raising the amphiphile content to 85 % by adding further amounts of amphiphile (200 ppm amphiphile, 35 ppm CFI) provided a slightly better WSD (as expected), but it did not provide any additional CFPP improvement.

Raising the amphiphile content of the additive mixture to 86 % by using less CFPP additive (20 ppm) did not show a comparable CFPP improvement, nor any other significantly beneficial effect.

Table B, which summarizes the results of the testing in Test oil 7, shows experimental data obtained with additive combinations containing a minor amount of amphiphile and a major amount of CFI. The results for both Polymer F and Polymer H show that additive combinations containing 78 % (350 ppm) of CFI and 22 % (100 ppm) of amphiphile 1 provide a CFPP improvement in comparison to the treatment with CFI alone. This CFPP improvement effect was observed only when terpolymers: Polymer F and Polymer H made of ethylene, vinyl acetate and the vinyl ester of a neocarboxylic acid were used, but the CFPP improvement did not occur with the conventional EVA copolymer A. Furthermore, the WSD was improved with the additive of the invention in comparison to the fuel containing the same amount of amphiphile 1 without any CFI.

Raising the CFI content of the additive mixture to 88 % by using less amphiphile (50 ppm) did not result in a CFPP improvement when compared to the addition of the CFI additive alone.

Raising the CFI content of the additive mixture to 86 % by using more CFPP additive (600 ppm) did not show any additional CFPP improvement in comparison to the fuel containing no amphiphile.

As shown by these experiments and the experiments presented in the subject Application, the combination of an amphiphile A) such as glycerol monooleate, oleic acid diethanolamide, oleic acid, tall oil fatty acid, polyisobutenylsuccinic anhydride diesterified with diethylene glycol, and C_{18}-H_{35}-O-CH_2-CH(OH)-CH_2OH, and a cold flow improver B) being a terpolymer comprising ethylene, vinyl acetate and vinyl esters of neocarboxylic acids - particularly neononanoate, neodecanoate, neoundecanoate, or neododedeconoate, when the mixing ratio of component A) and B) is between 80 : 20 and 20 : 80, surprisingly provided significantly better filterability than similar combinations of amphiphiles with ethylene copolymers without such vinyl esters of neocarboxylic acids.

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Table A: Tests results in test oil 6

		Cold flow improver, active ingredients						
Amphiphile		CFI „A" @35 ppm	CFI „A" @20 ppm	CFI „F" @ 35 ppm	CFI „F" @20 ppm	CFI „G" @35 ppm	CFI „G" @20 ppm	none
amphiphile 6 @120 ppm	WSD	399	395	386	396	382	402	400
	ADT	13,5	10,2	5,5	5,5	5,6	5,8	5,7
	CFPP	-18	-12	-20	-12	-21	-14	-9
amphiphile 6 @200 ppm	WSD	375	n. d.	358	n. d.	354	n. d.	376
	ADT	13,5	n. d.	6,5	n. d.	6,2	n. d.	6,1
	CFPP	-18	n. d.	-20	n. d.	-20	n. d.	-9
none	WSD	672	676	665	670	668	670	674
	ADT	13,2	9,7	5,6	5,4	5,6	5,4	5,4
	CFPP	-18	-12	-18	-12	-19	-14	-9

n. d. = not determined

Table B: Tests results in test oil 7

		Cold flow improver, active ingredients						
Amphiphile		CFI „A" @350 ppm	CFI „A" @600 ppm	CFI „F" @350 ppm	CFI „F" @600 ppm	CFI „H" @350 ppm	CFI „H" @600 ppm	none
amphiphile 1 @100 ppm	WSD	427	432	415	413	408	410	415
	ADT	32,6	43,5	9,5	10,2	10,0	10,6	4,9
	CFPP	-17	-19	-21	-21	-22	-23	-10
amphiphile 1 @50 ppm	WSD	622	n. d.	618	n. d.	612	n. d.	625
	ADT	36,3	n. d.	10,5	n. d.	9,1	n. d.	4,7
	CFPP	-17	n. d.	-19	n. d.	-20	n. d.	-10
none	WSD	706	692	701	677	698	675	724
	ADT	35,6	41	10,4	12,6	9,3	11,2	4,8
	CFPP	-17	-19	-19	-21	-20	-23	-10

n. d. = not determined

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Frankfurt am Main,

Date: 05.12.2006

Matthias Krull